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(54) **ALUMINUM NITRIDE SINTERED COMPACT AND ITS PRODUCTION**

(57)Abstract:
PURPOSE: To produce the title highly reliable AlN sintered compact with less variance in the mechanical strength and having a low probability of generating defective products by sintering a compact of specified AlN powder.
CONSTITUTION: From 2.5W15pts.wt. binder (e.g., PVA), 0.01W5pts.wt. deflocculant (e.g., glycerin trioleate), and 0.4W15pts.wt. plasticizer (e.g., dimethyl phthalate) are incorporated into 100pts.wt. AlN powder having 0.1W1.5μm mean primary particle diameter and contg. ≤10wt.% primary particles having ≥2.0μm diameter, ≤1.5wt.% oxygen, ≤0.3wt.% cationic impurities, and ≤0.1wt.% C. The mixture is compacted, the compact is heated at 300W1,100° C for 5W24hr to remove the binder and plasticizer, and an AlN powder compact is obtained. The compact is then sintered at 1,700W2,100° C in the nonoxidizing atmosphere at atmospheric pressure to obtain an AlN sintered compact contg. ≤0.5wt.% O2, ≤0.3wt.% cationic impurities, and ≤0.1wt.% C and having ≥3.20g/cm3 density and ≥17 Weibull coefficient.

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CLAIMS

[Claim(s)]
[Claim 1] The alumimium nitride sintered compact with which the cation impurity contained when an oxygen content sets an alumimium nitride presentation to AlN 0.5 or less % of the weight is characterized by 0.07 or less % of the weight and a consistency being [three or more 3.20 g/cm and Weibull modulus] 17 or more for a carbon content 0.3 or less % of the weight.
[Claim 2] The manufacture approach of an alumimium nitride sintered compact given in the 1st term of the range of the application for patent characterized by making the alumimium nitride powder Plastic solid whose cation impurity contained when the first [an average of] particle diameter mainly consists of alumimium nitride fine particles whose primary particle 2.0 micrometers or more is 10 or less % of the weight by 0.1-1.5 micrometers and an oxygen content sets an alumimium nitride presentation to AlN 1.5 or less % of the weight is 0.3 or less % of the weight, and whose carbon content is 0.07 or less % of the weight sinter.

[Translation done.]

* NOTICES *

DETAILED DESCRIPTION

[Detailed Description of the Invention]
(Field of the Invention)
This invention relates to a new alumimium nitride sintered compact especially an alumimium nitride sintered compact with large Weibull modulus, and its manufacture approach.
(Trouble which a Prior art and invention tend to solve)
Since the alumimium nitride sintered compact has properties, such as high thermal conductivity, corrosion resistance, high intensity, and electric insulation, it is matter which attracts attention as new materials. For example, an oxygen content is [0.8 or less % of the weight and a cation impurity] 0.3 or less % of the weight and a high grade, and, moreover, the alumimium nitride sintered compact indicated by JP,59-50078,A is a sintered compact of high density. For this reason, the above-mentioned alumimium nitride sintered compact is the ingredient which was excellent in a thermal property, chemical property, and a mechanical property, showed especially translucency and was excellent also in optical property.
By the way, generally the mechanical strength of a ceramic ingredient shows big variation. This is because a mechanical strength is determined by the weakest part that exists in a ceramic ingredient. The big variation to a mechanical strength also in an alumimium nitride sintered compact is seen. The variation in a mechanical strength can be expressed with Weibull modulus, and variation becomes small, so that Weibull modulus is large. Generally, although the Weibull modulus of an alumimium nitride sintered compact is called about ten ("ceramic Ingredient Technical Collection" industrial technical pin center,large issue), its probability of occurrence of the defective according to lack of a mechanical strength at a value of this level is high, and it is deficient in it in dependability.
According to the place which this invention persons measured, the Weibull modulus of an alumimium nitride sintered compact which has the property which was excellent in the above-mentioned official report shows 13. Although this value is excellent compared with the above-mentioned common alumimium nitride sintered compact, in order to reduce the probability of occurrence of the defective by lack of a mechanical strength, still, it is not a satisfying enough value.
(Means for solving a trouble)
this invention persons had the small variation in a mechanical strength, and probability of occurrence of a defective was low, and they repeated research for the purpose of obtaining a reliable alumimium nitride sintered compact. Consequently, when the first [an average of] particle diameter of the alumimium nitride fine particles used as a raw material uses small what has few coarse grain, it finds out that the alumimium nitride sintered compact which has the outstanding description which attained the above-mentioned purpose is obtained, and it came to propose this invention.
That is, this invention is an alumimium nitride sintered compact with which a carbon content is characterized by a consistency being [three or more 3.20 g/cm and Weibull modulus] 17 or more 0.07 or less % of the weight 0.3 or less % of the weight by the cation impurity contained when an oxygen content sets an alumimium nitride presentation to AlN 0.5 or less % of the weight.
In addition, the alumimium nitride in this invention means 1:1 compounds of aluminum and nitrogen, and treats all things other than this as an impurity. However, although the front face of alumimium nitride powder and a sintered compact oxidized unescapable in air and has placed and changed aluminum-N association to aluminum-O association, it is not considered that this association aluminum is a cation impurity. Therefore, the metal aluminum which has not carried out above-mentioned aluminum-N and aluminum-O association is a cation impurity.

An oxygen content has [0.5 or less % of the weight and a cation impurity / the content of an impurity] very few 0.3 or less % of the weight and carbon contents like 0.07 or less % of the weight, and, moreover, the sintered density of the alumimium nitride sintered compact of this invention is a precise sintered compact called three or more 3.20 g/cm. And the Weibull modulus which shows the variation in a mechanical strength shows the good value or more of 17.
The Weibull modulus in this invention is the value which asked for the three-point flexural strength about 50 test pieces according to the formula measured and mentioned later.
a grade with few contents of an impurity to the alumimium nitride sintered compact of this invention -- it is in the inclination which shows **** Weibull modulus. Here, it becomes an impurity from carbon as a cation about oxygen as an anion substantially again at the matter list which can become the cation and cation of a metal and others. The content of these impurities is lessened further, for example, an oxygen content can consider as the alumimium nitride sintered compact which has the more excellent Weibull modulus, for example, 18 or more Weibull moduli, when a cation impurity considers as 0.1 or less % of the weight and a carbon content ****s to 0.07 or less % of the weight 0.3 or less % of the weight. Also in the above-mentioned impurity, although it can consider that especially a carbon content is also a cation from the point which can be combined with an anion, about carbon, the content is specified separately and it is specified as 0.07 or less % of the weight, and the still fewer one is desirable, for example, it is still more suitable for it, since Weibull modulus is influenced greatly that it is 0.05 or less % of the weight 0.06 or less % of the weight.
The alumimium nitride sintered compact of this invention also combines and has the description [have the property which was excellent as mentioned above, and also] that surface roughness is small. A front face is ground in order for an alumimium nitride sintered compact to raise the smooth nature on the front face of a sintered compact after sintering generally. Like the alumimium nitride sintered compact of this invention, when surface roughness is small, the time amount which the surface polish after sintering takes can be omitted, and the surface polish itself can be omitted further. In Ra, the surface roughness of the non-polished surface of the alumimium nitride sintered compact of this invention is 1.0 micrometers or less, and is a value far smaller than 7.0 micrometers of a common alumimium nitride sintered compact.
Furthermore, since there are very few contents of an impurity, as for the alumimium nitride sintered compact of this invention, it has remarkable high translucency to the light - infrared light. For example, the thing used as the sintered compact which has the outstanding engine performance whose absorption coefficient to light with a wavelength of 6 micrometers is one or less [60cm -] in the formula of following Lambert-Beer also exists.
I=Ioe-mutIo: -- on-the-strength I: of incident light -- on-the-strength t: of the transmitted light -- thickness mu: of an ingredient -- absorption coefficient The alumimium nitride sintered compact which has the above outstanding properties is obtained only after it satisfies said requirements for the versatility carried out. That is, the requirements which are four whose cation impurities which the oxygen content in an alumimium nitride sintered compact contains at 0.5 or less % of the weight are 0.3 or less % of the weight, whose carbon contents are 0.07 or less % of the weight and, whose sintered density is three or more 3.20 g/cm cannot serve as alumimium nitride of this invention, even if the one requirement of which is missing. The amount of content oxygen has the outstanding property to which a content cation impurity says [Weibull modulus] 0.1 or less % of the weight, and 0.07 or less % of the weight and sintered density say [a carbon content] a three or more 3.22 g/cm alumimium nitride sintered compact or more as 18 0.3 or less % of the weight especially among the above-mentioned requirements.
The alumimium nitride sintered compact of this invention will not be limited especially if said requirements are satisfied regardless of the process. It will be as follows if the typical manufacture approach generally adopted suitably is illustrated.
It is the approach of making the alumimium nitride powder Plastic solid whose cation impurity contained when it mainly consists of alumimium nitride fine particles whose first [an average of] particle diameter is 0.1-1.5 micrometers, and whose primary particle 20 micrometers or more is 10 or less % of the weight and an oxygen content sets an alumimium nitride presentation to AlN 1.5 or less % of the weight is 0.3 or less % of the weight and whose carbon content is 0.07 or less % of the weight sintering.
Moreover, the first [an average of] particle diameter mainly consists of alumimium nitride fine particles whose primary particle (0.1-1.5 micrometers and 2.0 micrometers or more) is 10 or less % of the weight. therefore, this invention -- The cation impurity contained when an oxygen content sets an alumimium

nitride presentation to AlN 1.5 or less % of the weight Below 0.3% weight The oxygen content characterized by making the alumimium nitride powder Plastic solid whose carbon content is 0.1 or less % of the weight sinter 0.5 or less % of the weight, A carbon content also offers [the cation impurity contained when setting an alumimium nitride presentation to AlN] the manufacture approach of an alumimium nitride sintered compact that a consistency is [three or more 3.20 g/cm and Weibull modulus] 17 or more, 0.07 or less % of the weight 0.3 or less % of the weight.
The first [an average of] particle diameter is 0.1-1.5 micrometers, and the alumimium nitride powder Plastic solid with which sintering is presented mainly consists of alumimium nitride fine particles whose primary particle 2.0 micrometers or more is 10 or less % of the weight. In addition, the first [an average of] particle diameter is the value calculated from the electron microscope photograph so that it might explain in full detail behind. When the particle diameter of alumimium nitride fine particles has big effect on the Weibull modulus of the alumimium nitride sintered compact obtained and it separates from the above-mentioned range, the alumimium nitride sintered compact which has the property which was excellent in previous statement is not obtained. The first [an average of] above-mentioned particle diameter is 0.2-1.0 micrometers preferably. Moreover, as for a primary particle 2.0 micrometers or more, it is desirable that it is 3 or less % of the weight, and further completely not existing is more desirable.
The alumimium nitride powder Plastic solid with which sintering is presented must have very few contents of an impurity, the cation impurity contained when an oxygen content sets an alumimium nitride presentation to AlN 1.5 or less % of the weight must be 0.3 or less % of the weight, and a carbon content must be 0.07 or less % of the weight. The description of the alumimium nitride sintered compact obtained becomes good, so that there are few contents of these impurities. Therefore, the alumimium nitride powder Plastic solid 0.1 or less % of the weight and whose carbon content of 0.3 - 1.2 % of the weight and a cation impurity an oxygen content is 0.06 or less % of the weight is used preferably.
The higher one of the shaping consistency of the above-mentioned alumimium nitride powder Plastic solid is desirable, and when the description of the alumimium nitride sintered compact obtained is taken into consideration, it is desirable that it is three or more further 1.75 g/cm three or more 1.70 g/cm.
Although you may obtain by what kind of approach, the above-mentioned alumimium nitride powder Plastic solid is as follows when a suitable approach is mentioned.
(1) The approach a primary particle 2.0 micrometers or more is [the first / an average of / particle diameter] 10 or less % of the weight in 0.1-1.5 micrometers, and an oxygen content carries out pressing of the alumimium nitride fine particles 0.3 or less % of the weight and whose carbon content a cation impurity is 0.07 or less % of the weight by the pressure of 200-4000kg/cm2 with a pressurizer 1.5 or less % of the weight.
In the case of this approach, in order to make good the moldability of an alumimium nitride powder Plastic solid, a well-known additive can be added, but it must be the addition which becomes below the specific value that the oxygen content, the amount of cation impurities, and carbon content in the alumimium nitride powder Plastic solid acquired described above. In order to lessen the amount of the impurity in an alumimium nitride powder Plastic solid, fabricating without addition of an additive etc. only by pressurization is desirable.
(2) The approach an oxygen content carries out heating removal of this binder after 0.3 or less % of the weight and a carbon content fabricate the mixture which a primary particle 2.0 micrometers or more is 10 or less % of the weight in 0.1-1.5 micrometers, and the first [an average of] particle diameter added the binder to the alumimium nitride fine particles 1.5 or less % of the weight and whose cation impurity are 0.07 or less % of the weight, and was obtained.
As a binder which can be used in the approach of the above (2), the well-known compound currently used as a binder of ceramic fine particles can use it that there is no limit in any way. For example, the organic high molecular compound disassembled at the temperature of 1100 degrees C or less is adopted suitably. if the binder suitably used in this invention is illustrated -- oxygenated organic high molecular compound [, such as polyvinyl grumble RARU, polymethylmethacrylate, cellulose acetate butylate, a nitrocellulose, polyacrylic ester, polyvinyl alcohol, methyl cellulose, a hydroxymethyl cellulose, and polyethylene oxide]: -- in addition to this -- hydrocarbon system synthetic resin, such as petroleum resin, polyethylene, polypropylene, and polystyrene, etc. -- a kind -- or it is used by two or more sorts, mixing. the amount of such admixture used -- general -- the alumimium nitride fine-particles 100 weight section -- receiving -- 2.5 - 15 weight section -- what is necessary is just to choose from the range of 4 - 10 weight section preferably

Into the mixture of alumimium nitride fine particles and a binder, further, a deflocculatant and a plasticizer can be added in order to raise such dispersibility. As a deflocculatant, it is synthetic surface-active-agent; higher-fatty-acid; benzenesulfonic acid of a glycerol [of fatty acids, such as glycerol trio REETO and sorbitan trioleate,], or sorbitol ester; wild fish oil; non-ion system etc., for example. Moreover, as a plasticizer, they are stearic-acid-ester; tricresol phosphate; tree N-butyl phosphate; glycerols, such as phthalic ester; butyl stearate, such as a polyethylene glycol and its derivative; dimethyl phthalate, dibutyl phthalate, butyl benzyl phthalate, and dioctyl phthalate, etc., for example. In the addition of these deflocculatants and plasticizers, generally, a deflocculatant should just choose 0.01 - 5 weight section and a plasticizer from the range of 0.4 - 15 weight section to the alumimium nitride fine-particles 100 weight section.

Although a compound in which decomposition removal is carried out as an additive added to alumimium nitride fine particles by the heat-treatment mentioned later is permitted, it is not removed by heating, either and the compound which turns into a cation impurity or other impurities, and remains in an alumimium nitride powder Plastic solid is not suitable. Therefore, in this invention, addition of various kinds of metallic compounds known as sintering acid is not desirable.

As for mixing of the above-mentioned alumimium nitride fine particles, a binder and the deflocculatant added further as occasion demands, and a plasticizer, it is desirable to carry out wet blending in a nonaqueous solvent like alcohols, such as ketones; ethanol, such as an acetone and a methyl ethyl ketone, propanol, and a butanol.

In this way, the mixture of the alumimium nitride fine particles and the binder which were obtained is fabricated by the configuration of arbitration, such as the shape of a sheet, and tabular, with the sheet making machine and press-forming machine of a rubber press or a sheet forming method, for example, a doctor blade method. And decomposition removal of the binder is carried out by heating next. As for decomposition removal of the binder by heating, it is desirable to adopt the conditions which can also remove the carbon which a binder disassembles and generates. When the carbonaceous amount of survival increases and it exceeds 1.0 % of the weight, the alumimium nitride sintered compact of this invention may not be obtained. As for the conditions of heating, a 300-1100-degree C temperature requirement is suitably adopted as the bottom of oxygen, nitrogen-gas-atmosphere mind, or a vacuum, and in order for heating time to remove nearly completely the carbon generated by disassembly of a binder, the range of 5 - 24 hours is adopted suitably.

Although the alumimium nitride powder Plastic solid used by the approach of this invention is suitably manufactured by the approach described above, it is necessary to control it below to the specific value which described above the oxygen content contained in the alumimium nitride powder Plastic solid which is acquired in any case of an approach, the amount of cation impurities, and the carbon content.

Sintering is presented with an alumimium nitride powder Plastic solid next.

The method of calcinating sintering at an elevated temperature under a vacuum, the non-oxidizing atmosphere of atmospheric pressure, for example, ambient atmospheres, such as nitrogen gas, gaseous helium, and argon gas, or the nitrogen gas pressurization of 2 - 100 atmospheric-pressure extent is mentioned. The approach of calcinating especially under atmospheric pressure can adopt preferably. As a burning temperature, in the case of a vacuum or the non-oxidizing atmosphere of atmospheric pressure, the temperature of 1750-2050 degrees C is adopted suitably, and 1700-2100 degrees C of 1700-2400 degrees C of temperature of 1750-2300 degrees C are preferably adopted suitably under the nitrogen gas pressurization of two to 100 atmospheric pressure. In addition, the temperature in this invention is the value compensated so that the front face of the graphite crucible into which the alumimium nitride powder Plastic solid was put might be measured with a radiation thermometer and the gas temperature in a graphite crucible might be shown.

In this invention, in order to make high Weibull modulus of the alumimium nitride sintered compact obtained and to consider as a precise thing, it is desirable to make the average programming rate of an at least 1300-1700-degree C temperature requirement into the range of 1 degree C / min - 40 degrees C / min at the time of baking. It is more desirable to carry out a temperature up in the range of further 5-30 degrees C / min.

By the above manufacture approach, the alumimium nitride sintered compact of above-mentioned this invention is obtained.

(Effect **)

As stated above, the alumimium nitride sintered compact of this invention has high Weibull modulus, as

described above, the variation in a mechanical strength is very small, and its probability of occurrence of the defective by lack of a mechanical strength is very small. For example, when the defective probability of occurrence of Weibull modulus 17 of the alumimium nitride sintered compact of this invention and Weibull modulus 13 of an already well-known alumimium nitride sintered compact is compared, although the probability destroyed with 90% stress of the average reinforcement of the population about three-point flexural strength is about 8% in the former, it is about 16% in the latter. Therefore, it can be said that the alumimium nitride sintered compact of this invention is a ceramic ingredient which has high dependability. Of course, three-point flexural strength is a well-known alumimium nitride sintered compact and more than equivalent, and shows the good value or more [30kg //mm] of two.
Furthermore, the alumimium nitride sintered compact of this invention has the small surface roughness after sintering. Therefore, it is also possible to be able to shorten the time amount which grinds a front face and to omit the surface polish itself further after sintering, like the conventional alumimium nitride sintered compact.
And the alumimium nitride sintered compact of this invention is an epoch-making ingredient which has the large light transmission range in a light - infrared light field as described above further. Therefore, the alumimium nitride sintered compact of this invention is expected as a nitride ingredient with new hot aperture ingredient, light filter, frequency-conversion component, heat dissipation substrate of an integrated circuit, etc., and this industrial value is very large.
(Example)
Hereafter, although this invention is concretely illustrated according to an example, this invention is not limited to these examples.
In addition, in an example, the Weibull modulus of an alumimium nitride sintered compact and the first [an average of] particle diameter of alumimium nitride fine particles are the values calculated by the following approaches.
(1) Weibull modulus When the Weibull statistics display based on a weakest link theory shows the statistics display of the reinforcement of a ceramic ingredient, a Weibull destructive probability distribution function is given by the following formula.

$$\ell n \ell n \left(\frac{1}{1 - F_i} \right)$$

$$= m \ell n \sigma_R + m \ell n \left\{ \Gamma \left(\frac{m+1}{m} \right) / \mu \right\}$$

Fi: Probability of failure (accumulation probability)
When a strong ordinal number (number which set to i= 1 what has smallest sigmaR among the test piece, and gave sequential to ascending order) is set to i and the number of test pieces (measurement size) is set to N $\frac{i - 0.5}{N}$

since -- it computed.
m: Weibull-modulus sigmaR : maximum stress (reinforcement of a test piece)
mu: Average reinforcement It is based on the above-mentioned formula, $\ell n \ell n \left(\frac{1}{1 - F_i} \right)$

InsigmaR is plotted on an axis of abscissa and an axis of ordinate is asked for Weibull modulus from a slope of a line.
In addition, in this invention, about 50 test pieces, three-point flexural strength was measured according to JIS R-1601, and it asked for Weibull modulus from these data.
(2) First [an average of] particle diameter The photograph of 20 screens of the arbitration of AlN fine particles was taken by one 20000 times the scale factor of this with the scanning electron microscope. Parallel lines were drawn in the acquired photograph at 0.5-micrometer interval. Only the thing with the perfect profile was made into the object particle in the particle concerning parallel lines, and 500 or more

object particles were taken up out of 20 screens. The particle diameter of each particle was perpendicular to the parallel lines concerning each particle, and pulled two lines which turn into a particle tangent, and asked for them from the distance during the intersection of these lines and the above-mentioned parallel lines (l). In addition, when two or more lines started one particle, the distance during said intersection (l) counted only the greatest value. Moreover, when the whole particle diameter was quite large, the parallel lines of 2micro spacing were pulled in the 5000 times as many photograph as this, and same processing was carried out, and when still larger, by 2000 times, the parallel lines of 5micro spacing were pulled and it processed similarly.
First [an average of] weight criteria particle diameter and the diameter distribution of a primary particle were computed by the usual approach based on 500 or more obtained data.
Example 1 The dibutyl phthalate 12.2 weight section was mixed in the toluene-ethanol mixed solvent of 61 weight sections as a deflocculatant to the alumimium nitride powder 100 weight section shown in the 1st table by having used glycerol trio REETO as the 1.6 weight section and a plasticizer, having used the polyvinyl butyral as the 7.3 weight sections as a binder, and slurry was prepared. After fabricating this slurry in the shape of a sheet using the sheet making machine of a doctor blade method and fully drying, the test piece of 65mm angle was pierced. 600 degrees C of this test piece were heated with the small muffle furnace for 8 hours. The thickness of the acquired alumimium nitride powder Plastic solid was 1.19mm, and the shaping consistencies were 1.80 g/cm3, and chemical composition was as in the 1st table.
The acquired alumimium nitride powder Plastic solid was put into the crucible made from a graphite which carried out coating processing by boron nitride, and ordinary pressure sintering was carried out in an electric furnace and under nitrogen-gas-atmosphere mind. sintering -- 1850 degrees C from a room temperature -- until -- the temperature up of the programming rate was carried out by part for 10-degree-C/, and it carried out by holding at 1850 degrees C for 7 hours. In addition, the flow rate of nitrogen gas was considered as a part for 1**/. The obtained alumimium nitride sintered compact was light gray, and was what has translucency. Many of the physical properties were as being shown in the 2nd table.

第 1 表

諸物性		窒化アルミニウム粉体	窒化アルミニウム粉成形体
平均一次粒子径		0.5μm	0.5μm
2.0μm以上の一次粒子		なし	なし
比表面積		3.4㎡/g	3.4㎡/g
酸素含有量		0.9重量%	1.0重量%
炭素含有量		280ppm	430ppm
陽イオン不純物	Fe	7ppm	11ppm
	Si	43ppm	45ppm
	Ca	170ppm	150ppm
	Ni	5ppm以下	7ppm
	Cr	5ppm以下	5ppm

第　2　表

化学的性質	酸素含有量	0.28重量%
	炭素含有量	300ppm
	陽イオン不純物 Fe	15ppm
	Si	51ppm
	Ca	140ppm
	Ni	5ppm以下
	Cr	5ppm以下
物理的性質	密度	3.23g／cm³
	ワイブル係数	21
	曲げ強度	34kg／mm²
	未研磨状態の表面粗度	R_a =0.54μm
	6μ波長の光の吸収係数	43cm^{-1}

Example 2 Using various alumimium nitride fine particles, the alumimium nitride powder Plastic solid was created like the example 1, and it sintered after heat-treatment. The physical properties of the obtained alumimium nitride sintered compact were shown in the 3rd table.
In addition, the example performed like the above was shown in No.5 using the alumimium nitride fine particles indicated by JP,59-50078,A for the comparison. Furthermore, the example using alumimium nitride fine particles with much coarse grain was shown in No.6.

第　3　表

No.	窒化アルミニウム粉体 酸素含有量 (重量%)	表面積 (m²/g)	平均一次粒子径 (μm)	2μm以上の一次粒子の含有量(重量%)	窒化アルミニウム粉成形体 酸素含有量 (重量%)	炭素含有量 (ppm)	陽イオン不純物(ppm) Fe	Si	Ca	密度 (g/cm³)
1	0.9	3.3	0.5	0	1.0	390	<5	28	140	1.77
2	1.0	4.6	0.4	0	1.1	450	7	40	125	1.82
3	0.9	3.0	0.6	0	0.9	470	<5	39	170	1.78
4	1.2	3.5	0.7	1	1.4	350	50	95	110	1.88
5	1.0	3.7	0.7	12	1.2	400	10	73	95	1.92
6	0.4	0.7	5.0	80	0.50	280	27	65	100	1.95

No.	窒化アルミニウム焼結体									
	酸素含有量 (重量%)	炭素含有量 (ppm)	陽イオン不純物(ppm)			密度 (g/cm³)	6μ光吸収係数 (cm⁻¹)	ワイブル係数	曲げ強度 (kg/mm²)	未研磨面表面粗度 Ra(μm)
			Fe	Si	Ca					
1	0.25	280	11	30	135	3.23	45	19	33	0.61
2	0.21	410	13	38	110	3.22	42	18	37	0.48
3	0.30	320	8	45	150	3.22	46	20	35	0.52
4	0.41	270	45	105	90	3.20	58	17	30	0.74
5	0.39	330	12	78	100	2.78	測定不能	11	21	1.7
6	0.45	250	25	75	93	2.36	〃	9	16	3.2

但し、No.5及びNo.6は比較例である。

Example 3 The alumimium nitride sintered compact was obtained like the example 1 except having changed the addition of a binder, and the conditions of heat-treatment, as shown in the 4th table. The result was shown in the 4th table.

第 4 表

No.	成形体中の結合剤* (重量部)	加熱処理			窒化アルミニウム粉成形体					
		温度 (℃)	時間 (時間)	平均昇温速度 (℃/分)	酸素含有量 (重量%)	炭素含有量 (ppm)	陽イオン不純物(ppm)			密度 (g/cm³)
							Fe	Si	Ca	
1	15	600	7	0.5	1.0	450	11	48	148	1.79
2	15	350	20	0.3	1.0	510	9	51	145	1.82
3	18	1100	10	0.5	0.9	570	10	43	153	1.78
4	20	600	3	0.3	0.9	1050	10	47	147	1.83
5	25	600	4	0.5	1.0	1270	9	50	160	1.80

No.	窒化アルミニウム焼結体									
	酸素含有量 (重量%)	炭素含有量 (ppm)	陽イオン不純物(ppm)			密度 (g/cm³)	6μ光吸収係数 (cm⁻¹)	ワイブル係数	曲げ強度 (kg/mm²)	未研磨面表面粗度 Ra(μm)
			Fe	Si	Ca					
1	0.25	310	13	43	130	3.22	48	18	35	0.59
2	0.21	420	15	57	135	3.23	45	22	31	0.58
3	0.29	370	11	52	147	3.23	49	20	32	0.63
4	0.18	920	11	44	142	2.81	測定不能	12	13	1.15
5	0.17	1100	17	51	149	2.64	〃	10	15	1.28

* 窒化アルミニウム粉体100重量部に対する添加割合
No.4及びNo.5は比較例である。

Example 4 About 1.5g of alumimium nitride powder of an example 1 was put into the metal mold of 20mmphi, it preformed by 200kg/cm2, and, subsequently isostatic press shaping between the colds of this was carried out by the pressure of 3000kg/cm2. The consistencies of the acquired Plastic solid were 1.82 g/cm3. Ordinary pressure sintering of this Plastic solid was carried out by the same approach as an example 1. The obtained sintered compact was a light gray light transmission body. The physical properties of this sintered compact were as in the 5th table.

第　5　表

化学的性質	酸素含有量		0.23重量%
	炭素含有量		430ppm
	陽イオン不純物	Fe	12ppm
		Si	53ppm
		Ca	170ppm
		Ni	5ppm以下
		Cr	5ppm以下
物理的性質	密度		3.23g／cm³
	ワイブル係数		19
	曲げ強度		31kg／mm²
	未研磨状態の表面粗度		R_a=0.63μm
	6μ波長の光の吸収係数		47cm^{-1}

Example of a comparison The alumimium nitride sintered compact was obtained like the example 1 to the alumimium nitride fine particles used in the example 1 except having carried out addition mixing of the CaO powder 3% of the weight as sintering acid. The result was shown in the 6th table.

第　6　表

化学的性質	酸素含有量		0.13重量%
	炭素含有量		370ppm
	陽イオン不純物	Fe	14ppm
		Si	43ppm
		Ca	0.4重量%
		Ni	5ppm以下
		Cr	5ppm以下
物理的性質	密度		3.23g／cm³
	ワイブル係数		14
	曲げ強度		29kg／mm²
	未研磨状態の表面粗度		R_a=6.9μm
	6μ波長の光の吸収係数		41cm^{-1}

[Translation done.]

TECHNICAL FIELD

(Field of the Invention)
This invention relates to a new alumimium nitride sintered compact especially an alumimium nitride sintered compact with large Weibull modulus, and its manufacture approach.

[Translation done.]

TECHNICAL PROBLEM

(Trouble which a Prior art and invention tend to solve)
Since the alumimium nitride sintered compact has properties, such as high thermal conductivity, corrosion resistance, high intensity, and electric insulation, it is matter which attracts attention as new materials. For example, an oxygen content is [0.8 or less % of the weight and a cation impurity] 0.3 or less % of the weight and a high grade, and, moreover, the alumimium nitride sintered compact indicated by JP,59-50078,A is a sintered compact of high density. For this reason, the above-mentioned alumimium nitride sintered compact is the ingredient which was excellent in a thermal property, chemical property, and a mechanical property, showed especially translucency and was excellent also in optical property.
By the way, generally the mechanical strength of a ceramic ingredient shows big variation. This is because a mechanical strength is determined by the weakest part that exists in a ceramic ingredient. The big variation to a mechanical strength also in an alumimium nitride sintered compact is seen. The variation in a mechanical strength can be expressed with Weibull modulus, and variation becomes small, so that Weibull modulus is large. Generally, although the Weibull modulus of an alumimium nitride sintered compact is called about ten ("ceramic Ingredient Technical Collection" industrial technical pin center,large issue), its probability of occurrence of the defective according to lack of a mechanical strength at a value of this level is high, and it is deficient in it in dependability.
According to the place which this invention persons measured, the Weibull modulus of an alumimium nitride sintered compact which has the property which was excellent in the above-mentioned official report shows 13. Although this value is excellent compared with the above-mentioned common alumimium nitride sintered compact, in order to reduce the probability of occurrence of the defective by lack of a mechanical strength, still, it is not a satisfying enough value.

[Translation done.]

MEANS

(Means for solving a trouble)
this invention persons had the small variation in a mechanical strength, and probability of occurrence of a defective was low, and they repeated research for the purpose of obtaining a reliable alumimium nitride sintered compact. Consequently, when the first [an average of] particle diameter of the alumimium nitride fine particles used as a raw material uses small what has few coarse grain, it finds out that the alumimium nitride sintered compact which has the outstanding description which attained the above-mentioned purpose is obtained, and it came to propose this invention.
That is, this invention is an alumimium nitride sintered compact with which a carbon content is characterized by a consistency being [three or more 3.20 g/cm and Weibull modulus] 17 or more 0.07 or less % of the weight 0.3 or less % of the weight by the cation impurity contained when an oxygen content sets an alumimium nitride presentation to AlN 0.5 or less % of the weight.
In addition, the alumimium nitride in this invention means 1:1 compounds of aluminum and nitrogen, and treats all things other than this as an impurity. However, although the front face of alumimium nitride powder and a sintered compact oxidized unescapable in air and has placed and changed aluminum-N association to aluminum-O association, it is not considered that this association aluminum is a cation impurity. Therefore, the metal aluminum which has not carried out above-mentioned aluminum-N and aluminum-O association is a cation impurity.
An oxygen content has [0.5 or less % of the weight and a cation impurity / the content of an impurity] very few 0.3 or less % of the weight and carbon contents like 0.07 or less % of the weight, and, moreover, the sintered density of the alumimium nitride sintered compact of this invention is a precise sintered compact called three or more 3.20 g/cm. And the Weibull modulus which shows the variation in a mechanical strength shows the good value or more of 17.
The Weibull modulus in this invention is the value which asked for the three-point flexural strength about 50 test pieces according to the formula measured and mentioned later.
a grade with few contents of an impurity to the alumimium nitride sintered compact of this invention -- it is in the inclination which shows **** Weibull modulus. Here, it becomes an impurity from carbon as a cation about oxygen as an anion substantially again at the matter list which can become the cation and cation of a metal and others. The content of these impurities is lessened further, for example, an oxygen content can consider as the alumimium nitride sintered compact which has the more excellent Weibull modulus, for example, 18 or more Weibull moduli, when a cation impurity considers as 0.1 or less % of the weight and a carbon content ****s to 0.07 or less % of the weight 0.3 or less % of the weight. Also in the above-mentioned impurity, although it can consider that especially a carbon content is also a cation from the point which can be combined with an anion, about carbon, the content is specified separately and it is specified as 0.07 or less % of the weight, and the still fewer one is desirable, for example, it is still more suitable for it, since Weibull modulus is influenced greatly that it is 0.05 or less % of the weight 0.06 or less % of the weight.
The alumimium nitride sintered compact of this invention also combines and has the description [have the property which was excellent as mentioned above, and also] that surface roughness is small. A front face is ground in order for an alumimium nitride sintered compact to raise the smooth nature on the front face of a sintered compact after sintering generally. Like the alumimium nitride sintered compact of this invention, when surface roughness is small, the time amount which the surface polish after sintering takes can be omitted, and the surface polish itself can be omitted further. In Ra, the surface roughness of the non-polished surface of the alumimium nitride sintered compact of this invention is 1.0

micrometers or less, and is a value far smaller than 7.0 micrometers of a common alumimium nitride sintered compact.
Furthermore, since there are very few contents of an impurity, as for the alumimium nitride sintered compact of this invention, it has remarkable high translucency to the light - infrared light. For example, the thing used as the sintered compact which has the outstanding engine performance whose absorption coefficient to light with a wavelength of 6 micrometers is one or less [60cm -] in the formula of following Lambert-Beer also exists.
I=Ioe-mutIo: -- on-the-strength I: of incident light -- on-the-strength t: of the transmitted light -- thickness mu: of an ingredient -- absorption coefficient The alumimium nitride sintered compact which has the above outstanding properties is obtained only after it satisfies said requirements for the versatility carried out. That is, the requirements which are four whose cation impurities which the oxygen content in an alumimium nitride sintered compact contains at 0.5 or less % of the weight are 0.3 or less % of the weight, whose carbon contents are 0.07 or less % of the weight and, whose sintered density is three or more 3.20 g/cm cannot serve as alumimium nitride of this invention, even if the one requirement of which is missing. The amount of content oxygen has the outstanding property to which a content cation impurity says [Weibull modulus] 0.1 or less % of the weight, and 0.07 or less % of the weight and sintered density say [a carbon content] a three or more 3.22 g/cm alumimium nitride sintered compact or more as 18 0.3 or less % of the weight especially among the above-mentioned requirements.
The alumimium nitride sintered compact of this invention will not be limited especially if said requirements are satisfied regardless of the process. It will be as follows if the typical manufacture approach generally adopted suitably is illustrated.
It is the approach of making the alumimium nitride powder Plastic solid whose cation impurity contained when it mainly consists of alumimium nitride fine particles whose first [an average of] particle diameter is 0.1-1.5 micrometers, and whose primary particle 20 micrometers or more is 10 or less % of the weight and an oxygen content sets an alumimium nitride presentation to AlN 1.5 or less % of the weight is 0.3 or less % of the weight and whose carbon content is 0.07 or less % of the weight sintering.
Moreover, the first [an average of] particle diameter mainly consists of alumimium nitride fine particles whose primary particle (0.1-1.5 micrometers and 2.0 micrometers or more) is 10 or less % of the weight. therefore, this invention -- The cation impurity contained when an oxygen content sets an alumimium nitride presentation to AlN 1.5 or less % of the weight Below 0.3% weight The oxygen content characterized by making the alumimium nitride powder Plastic solid whose carbon content is 0.1 or less % of the weight sinter 0.5 or less % of the weight, A carbon content also offers [the cation impurity contained when setting an alumimium nitride presentation to AlN] the manufacture approach of an alumimium nitride sintered compact that a consistency is [three or more 3.20 g/cm and Weibull modulus] 17 or more, 0.07 or less % of the weight 0.3 or less % of the weight.
The first [an average of] particle diameter is 0.1-1.5 micrometers, and the alumimium nitride powder Plastic solid with which sintering is presented mainly consists of alumimium nitride fine particles whose primary particle 2.0 micrometers or more is 10 or less % of the weight. In addition, the first [an average of] particle diameter is the value calculated from the electron microscope photograph so that it might explain in full detail behind. When the particle diameter of alumimium nitride fine particles has big effect on the Weibull modulus of the alumimium nitride sintered compact obtained and it separates from the above-mentioned range, the alumimium nitride sintered compact which has the property which was excellent in previous statement is not obtained. The first [an average of] above-mentioned particle diameter is 0.2-1.0 micrometers preferably. Moreover, as for a primary particle 2.0 micrometers or more, it is desirable that it is 3 or less % of the weight, and further completely not existing is more desirable.
The alumimium nitride powder Plastic solid with which sintering is presented must have very few contents of an impurity, the cation impurity contained when an oxygen content sets an alumimium nitride presentation to AlN 1.5 or less % of the weight must be 0.3 or less % of the weight, and a carbon content must be 0.07 or less % of the weight. The description of the alumimium nitride sintered compact obtained becomes good, so that there are few contents of these impurities.

EXAMPLE

(Example)
Hereafter, although this invention is concretely illustrated according to an example, this invention is not limited to these examples.
In addition, in an example, the Weibull modulus of an alumimium nitride sintered compact and the first [an average of] particle diameter of alumimium nitride fine particles are the values calculated by the following approaches.
(1) Weibull modulus When the Weibull statistics display based on a weakest link theory shows the statistics display of the reinforcement of a ceramic ingredient, a Weibull destructive probability distribution function is given by the following formula.

$$\ell_n \ell_n \left(\frac{1}{1 - F_i} \right)$$

$$= m \ell_n \sigma_R + m \ell_n \left\{ \Gamma \left(\frac{m + 1}{m} \right) / \mu \right\}$$

Fi: Probability of failure (accumulation probability)
When a strong ordinal number (number which set to i= 1 what has smallest sigmaR among the test piece, and gave sequential to ascending order) is set to i and the number of test pieces (measurement size) is

set to N $\frac{i - 0.5}{N}$

since -- it computed.
m: Weibull-modulus sigmaR : maximum stress (reinforcement of a test piece)

mu: Average reinforcement It is based on the above-mentioned formula, $\ell_n \ell_n \left(\frac{1}{1 - F_i} \right)$

InsigmaR is plotted on an axis of abscissa and an axis of ordinate is asked for Weibull modulus from a slope of a line.
In addition, in this invention, about 50 test pieces, three-point flexural strength was measured according to JIS R-1601, and it asked for Weibull modulus from these data.
(2) First [an average of] particle diameter The photograph of 20 screens of the arbitration of AlN fine particles was taken by one 20000 times the scale factor of this with the scanning electron microscope. Parallel lines were drawn in the acquired photograph at 0.5-micrometer interval. Only the thing with the perfect profile was made into the object particle in the particle concerning parallel lines, and 500 or more object particles were taken up out of 20 screens. The particle diameter of each particle was perpendicular to the parallel lines concerning each particle, and pulled two lines which turn into a particle tangent, and asked for them from the distance during the intersection of these lines and the above-mentioned parallel lines (l). In addition, when two or more lines started one particle, the distance during said intersection (l) counted only the greatest value. Moreover, when the whole particle diameter was

quite large, the parallel lines of 2micro spacing were pulled in the 5000 times as many photograph as this, and same processing was carried out, and when still larger, by 2000 times, the parallel lines of 5micro spacing were pulled and it processed similarly.
First [an average of] weight criteria particle diameter and the diameter distribution of a primary particle were computed by the usual approach based on 500 or more obtained data.
Example 1 The dibutyl phthalate 12.2 weight section was mixed in the toluene-ethanol mixed solvent of 61 weight sections as a deflocculatant to the alumimium nitride powder 100 weight section shown in the 1st table by having used glycerol trio REETO as the 1.6 weight section and a plasticizer, having used the polyvinyl butyral as the 7.3 weight sections as a binder, and slurry was prepared. After fabricating this slurry in the shape of a sheet using the sheet making machine of a doctor blade method and fully drying, the test piece of 65mm angle was pierced. 600 degrees C of this test piece were heated with the small muffle furnace for 8 hours. The thickness of the acquired alumimium nitride powder Plastic solid was 1.19mm, and the shaping consistencies were 1.80 g/cm3, and chemical composition was as in the 1st table.
The acquired alumimium nitride powder Plastic solid was put into the crucible made from a graphite which carried out coating processing by boron nitride, and ordinary pressure sintering was carried out in an electric furnace and under nitrogen-gas-atmosphere mind. sintering -- 1850 degrees C from a room temperature -- until -- the temperature up of the programming rate was carried out by part for 10-degree-C/, and it carried out by holding at 1850 degrees C for 7 hours. In addition, the flow rate of nitrogen gas was considered as a part for 1**/. The obtained alumimium nitride sintered compact was light gray, and was what has translucency. Many of the physical properties were as being shown in the 2nd table.

第　1　表

諸物性		窒化アルミニウム粉体	窒化アルミニウム粉成形体
平均一次粒子径		0.5μm	0.5μm
2.0μm以上の一次粒子		なし	なし
比表面積		3.4㎡/g	3.4㎡/g
酸素含有量		0.9重量%	1.0重量%
炭素含有量		280ppm	430ppm
陽イオン不純物	Fe	7ppm	11ppm
	Si	43ppm	45ppm
	Ca	170ppm	150ppm
	Ni	5ppm以下	7ppm
	Cr	5ppm以下	5ppm

第　2　表

化学的性質	酸素含有量		0.28重量%
	炭素含有量		300ppm
	陽イオン不純物	Fe	15ppm
		Si	51ppm
		Ca	140ppm
		Ni	5ppm以下
		Cr	5ppm以下
物理的性質	密度		3.23g/㎤
	ワイブル係数		21

	曲げ強度	34kg/mm^2
	未研磨状態の表面粗度	R_a =0.54μm
	6μ波長の光の吸収係数	43cm^{-1}

Example 2 Using various alumimium nitride fine particles, the alumimium nitride powder Plastic solid was created like the example 1, and it sintered after heat-treatment. The physical properties of the obtained alumimium nitride sintered compact were shown in the 3rd table.
In addition, the example performed like the above was shown in No.5 using the alumimium nitride fine particles indicated by JP,59-50078,A for the comparison. Furthermore, the example using alumimium nitride fine particles with much coarse grain was shown in No.6.

第 3 表

No.	窒化アルミニウム粉体				窒化アルミニウム粉成形体					
	酸素含有量(重量%)	表面積(m^2/g)	平均一次粒子径(μm)	2μm以上の一次粒子の含有量(重量%)	酸素含有量(重量%)	炭素含有量(ppm)	陽イオン不純物(ppm) Fe	Si	Ca	密度(g/cm^3)
1	0.9	3.3	0.5	0	1.0	390	<5	28	140	1.77
2	1.0	4.6	0.4	0	1.1	450	7	40	125	1.82
3	0.9	3.0	0.6	0	0.9	470	<5	39	170	1.78
4	1.2	3.5	0.7	1	1.4	350	50	95	110	1.88
5	1.0	3.7	0.7	12	1.2	400	10	73	95	1.92
6	0.4	0.7	5.0	80	0.50	280	27	65	100	1.95

No.	窒化アルミニウム焼結体									
	酸素含有量(重量%)	炭素含有量(ppm)	陽イオン不純物(ppm) Fe	Si	Ca	密度(g/cm^3)	6μ光吸収係数(cm^{-1})	ワイブル係数	曲げ強度(kg/mm^2)	未研磨面表面粗度 R_a(μm)
1	0.25	280	11	30	135	3.23	45	19	33	0.61
2	0.21	410	13	38	110	3.22	42	18	37	0.48
3	0.30	320	8	45	150	3.22	46	20	35	0.52
4	0.41	270	45	105	90	3.20	58	17	30	0.74
5	0.39	330	12	78	100	2.78	測定不能	11	21	1.7
6	0.45	250	25	75	93	2.36	〃	9	16	3.2

但し、No.5及びNo.6は比較例である。

Example 3 The alumimium nitride sintered compact was obtained like the example 1 except having changed the addition of a binder, and the conditions of heat-treatment, as shown in the 4th table. The result was shown in the 4th table.

第　　4　　表

No	成形体中の結合剤*	加熱処理			窒化アルミニウム粉成形体					
		温度	時間	平均昇温速度	酸素含有量	炭素含有量	陽イオン不純物(ppm)			密度
	(重量部)	(℃)	(時間)	(℃/分)	(重量%)	(ppm)	Fe	Si	Ca	(g/cm³)
1	15	600	7	0.5	1.0	450	11	48	148	1.79
2	15	350	20	0.3	1.0	510	9	51	145	1.82
3	18	1100	10	0.5	0.9	570	10	43	153	1.78
4	20	600	3	0.3	0.9	1050	10	47	147	1.83
5	25	600	4	0.5	1.0	1270	9	50	160	1.80

No	窒化アルミニウム焼結体									
	酸素含有量	炭素含有量	陽イオン不純物(ppm)			密度	6μ光吸収係数	ワイブル係数	曲げ強度	未研磨面表面粗度
	(重量%)	(ppm)	Fe	Si	Ca	(g/cm³)	(cm⁻¹)		(kg/mm²)	Ra(μm)
1	0.25	310	13	43	130	3.22	48	18	35	0.59
2	0.21	420	15	57	135	3.23	45	22	31	0.58
3	0.29	370	11	52	147	3.23	49	20	32	0.63
4	0.18	920	11	44	142	2.81	測定不能	12	13	1.15
5	0.17	1100	17	51	149	2.64	〃	10	15	1.28

＊ 窒化アルミニウム粉体100重量部に対する添加割合
No4及びNo5は比較例である。

Example 4 About 1.5g of alumimium nitride powder of an example 1 was put into the metal mold of 20mmphi, it preformed by 200kg/cm2, and, subsequently isostatic press shaping between the colds of this was carried out by the pressure of 3000kg/cm2. The consistencies of the acquired Plastic solid were 1.82 g/cm3. Ordinary pressure sintering of this Plastic solid was carried out by the same approach as an example 1. The obtained sintered compact was a light gray light transmission body. The physical properties of this sintered compact were as in the 5th table.

第　　5　　表

化学的性質	酸素含有量		0.23重量%
	炭素含有量		430ppm
	陽イオン不純物	Fe	12ppm
		Si	53ppm
		Ca	170ppm
		Ni	5ppm以下
		Cr	5ppm以下
物理的性質	密度		3.23g/cm³
	ワイブル係数		19
	曲げ強度		31kg/mm²
	未研磨状態の表面粗度		Ra＝0.63μm
	6μ波長の光の吸収係数		47cm⁻¹

Example of a comparison The alumimium nitride sintered compact was obtained like the example 1 to the alumimium nitride fine particles used in the example 1 except having carried out addition mixing of the

CaO powder 3% of the weight as sintering acid. The result was shown in the 6th table.

第　6　表

化学的性質	酸素含有量		0.13重量%
	炭素含有量		370ppm
	陽イオン不純物	Fe	14ppm
		Si	43ppm
		Ca	0.4重量%
		Ni	5ppm以下
		Cr	5ppm以下
物理的性質	密度		3.23g／cm³
	ワイブル係数		14
	曲げ強度		29kg／mm²
	未研磨状態の表面粗度		R_a＝6.9μm
	6μ波長の光の吸収係数		41cm^{-1}

[Translation done.]